RADIONUCLIDE CHARACTERIZATION AND ASSOCIATED DOSE FROM LONG-LIVED RADIONUCLIDES IN CLOSE-IN FALLOUT DELIVERED TO THE MARINE ENVIRONMENT AT BIKINI AND ENEWETAK ATOLLS

William L. Robison and Victor E. Noshkin
Lawrence Livermore National Laboratory (LLNL), Livermore, California USA

Enewetak and Bikini Atolls are located about 11º N and 162º and 165º E, respectively, in the Marshall Islands. These two sites in the northern Marshall Islands were used by the United States as testing grounds for nuclear devices beginning in 1946. At Enewetak, 19 of the 43 tests were made from barges anchored in the lagoon. The remaining tests included 2 air drops, 2 underwater tests, 7 ground surface tests and 13 tests with devices fixed to towers. Bikini Atoll was the first U.S. nuclear test site in the Pacific, and most of the 23 tests conducted at Bikini were detonated on barges anchored in the lagoon or on the reef. Two tests were air drops, one was underwater and three were ground surface explosions. This testing produced close-in fallout debris that was contaminated with quantities of radioactive fission and particle activated products, and unspent radioactive nuclear fuel that entered the aquatic environment of the atolls. The radionuclides deposited to the lagoon water either settled rapidly to the bottom sediments or remained as dissolved or particulate-associated species in the water and were eventually discharged to the ocean. The U.S. moratorium began on 31 October 1958 and marked an end of all nuclear testing at the atolls.

The lagoon sediments of Bikini an Enewetak lagoon are found at a mean depth of 46 ± 1 meter below the water surface. These exposed deposits at Bikini and Enewetak currently are a reservoir for more than 100 and 50 TBq, respectively, of Plutonium-293+240 ($^{239+240}$Pu) with lesser amounts of Americium-241 (^{241}Am), Plutonium-238 (^{238}Pu) and other long-lived fission and activation products such as Strontium-90 (^{90}Sr), Cesium-137 (^{137}Cs), and Bismuth-207 (^{207}Bi) to name a few. Radionuclides with half-lives less than 6–10 years such as Cobalt-60 (^{60}Co), Rhodium-101,102m (^{101}, ^{102m}Rh), Antimony-125 (^{125}Sb), and Europium-155 (^{155}Eu) have essentially disappeared from the sedimentary environment of the lagoon through radioactive decay. The concentration of the long-lived radionuclides varies around the lagoon, but generally highest levels are associated with fine and coarse carbonate materials adjacent to the locations of the larger explosions. The radionuclides are also distributed vertically in the sediment column to various depths and degree in all regions of the lagoon.

$^{239+240}$Pu, ^{241}Am, ^{90}Sr, ^{137}Cs and other fission and activation products at greater than fallout background concentrations are also found in the water sampled from all locations throughout the lagoons during all periods sampled between 1972 and 1995. This is a direct indication that the radionuclides are continuously mobilized to solution from the solid phases in these environments. For example, the variations between the average soluble concentrations of $^{239+240}$Pu determined from samples obtained during a 20 year period, are not at this time considered significant. The assumption is made that the standing average amount of plutonium in the lagoon water mass at any time, is constant. Stated differently, steady state conditions have been established for $^{239+240}$Pu partitioning from the sedimentary reservoirs to solution. These quantities in the lagoon water mass represent <0.1% of the sediment inventories determined to a depth of 16 cm at each atoll. Mobilization of $^{239+240}$Pu from marine deposits is a slow, but nevertheless real process.

The mobilized $^{239+240}$Pu at Enewetak and Bikini has solute-like characteristics and different valence states coexist in solution. The largest fraction of the soluble plutonium is in an oxidized form (+V or VI). Quantities associated with suspended particulate material and

sediments are predominately in the reduced state (+III or IV). The sorption-desorption process is not completely reversible because of changes that occur in the relative amounts of the mixed oxidation states in solution with time. The oxidized forms of $^{239+240}$Pu in solution have a lesser tendency to associate with sedimentary or particulate material than reduced plutonium. Complexation after mobilization also affects the resorption rate.

Also of particular importance is the fact that these long-lived radionuclides are accumulated to different levels by indigenous aquatic plants and organisms that may be used as food by people. Hence a major effort was devoted to dissections and analysis of the edible muscle tissue from a variety of fish [1]. Other studies were made to evaluate the variability of radionuclides in families of fish, define the major tissues or organs where radionuclides were concentrated by fish, develop concentration factors, and assess the effective half time for some of the radionuclides using the resident non-migratory reef fish as indicators of environmental change.

The estimated radiological dose from consumption of marine foods at Enewetak and Bikini Atolls is less than 0.1% of the total estimated 30 year integral effective dose from all exposure pathways [2,3]. The large existing inventories of $^{239+240}$Pu and ^{241}Am were directly dispersed into the marine environment and not encased in drums, glass or any other form of containment. In spite of these large, dispersed, exposed transuranic radionuclide inventories, they contribute very little to the total estimated dose from marine foods. The total radiological dose from the marine pathway is dominated by the natural radionuclides Polonium-210 (^{210}Po) and Lead-210 (^{210}Pb). The contribution of man-made, bomb-related radionuclides (^{137}Cs + $^{239+240}$Pu) is about 0.2% of the total dose estimated for the marine food chain(Table 1).

Table 1. Comparative dose from naturally occurring ^{210}Po/^{210}Pb and bomb related $^{239+240}$Pu and ^{137}Cs from fish from Enewetak lagoon.[a]

Natural ^{210}Po	0.26 mSv over 50 y
Natural ^{210}Pb	0.029 mSv over 50 y
Total ^{210}Po + ^{210}Pb	~0.29 mSv over 50 y
Bomb related $^{239+240}$Pu[b]	0.000045 mSv over 50 y
Bomb related ^{137}Cs	0.005 mSv over 50 y

[a] Based on the same intake of fish for both ^{210}Po and $^{239+240}$Pu calculations and the latest ICRP dose conversion factors.

[b] Based on a gut-transfer factor of 5×10^{-4}.

References:

[1] NOSHKIN, VE, ROBISON, W.L., WONG, K.M., JOKELA, T.A., and JONES, H.E. "Past and present levels of some radionuclides in fish from Bikini and Enewetak Atolls," *Health Physics* **73**(1), pp. 49–65 (1997).

[2] ROBISON, WL, CONRADO, C.L., and PHILLIPS, W.A., Enjebi Island Dose Assessment, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-53805 (1987).

[3] ROBISON, WL, BOGEN, K.T. and CONRADO, C.L., 'An updated dose assessment for resettlement options at Bikini Atoll—a U.S. nuclear test site," *Health Physics* **73**(1), (1997) 100–114.

Work performed under the auspices of the U.S. Department of Energy at Lawrence Livermore National Laboratory under contract W-7405-Eng-48.